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Abstract

The problem considered is the behavior of a gas bubble in a liquid saturated with dissolved gas when oscillating pressures are imposed on the system. This situation is encountered in experiments on cavitation and in the propagation of sonic and ultrasonic waves in liquids. Since gas diffuses into the bubble during the expansion half-cycle in which the pressure drops below its mean value, and diffuses out of the bubble during the compression half-cycle in which the pressure rises above its mean value, there is no net transfer of mass into or out of the bubble in first order. There is, however, in second order a net inflow of gas into the bubble which is called rectified diffusion. The equations which determine the system include the equation of state of the gas in the bubble, the equation of motion for the bubble boundary in the liquid, and the equation for the diffusion of dissolved gas in the liquid. In the solution presented here, the acoustic approximation is made; that is, the amplitude of the pressure oscillation is taken to be small. It is also assumed that the gas in the bubble remains isothermal throughout the oscillations; this assumption is valid provided the oscillation frequency is not too high. Under these conditions one finds for the mean rate of gas flow into the bubble the expression

$$\overline{(dm/dt)} = (8\pi/3) D C_{\infty} R_0 (\Delta P/P_0)^2$$

where D is the diffusivity of the dissolved gas in the liquid, C_{∞} is the equilibrium dissolved gas concentration for the mean ambient pressure P_0 , R_0 is the mean radius of the bubble, and ΔP is the amplitude of the acoustic pressure oscillations. It may be remarked that the most important contribution to the rectification effect comes from the convection contribution to the diffusion process.

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Introduction

When a gas bubble in a liquid saturated with dissolved gas is subject to an oscillating pressure field, a net flow of gas into the bubble over any complete cycle of oscillation is to be expected. An intuitive physical explanation is as follows. When the pressure rises above its mean value, the gas bubble is compressed and at the same time the concentration of dissolved gas in the liquid is below the equilibrium value appropriate for the increased pressure. This situation results in an outflow of gas from the bubble. On the other hand, by a corresponding argument, gas flows into the bubble from the liquid in its neighborhood during the expansion half-cycle. Because of the difference in surface area of the bubble between these two half-cycles, there will be a net gas flow into the bubble over a complete cycle. While the effect appears obvious from this intuitive argument, a quantitative analysis is not quite simple. First of all, the analytic difficulties are closely associated with the complicated dynamic problem which determines the motion of the bubble wall in terms of the applied pressure and other relevant parameters of the bubble. Secondly, even when the motion of the bubble wall is given, one still has to treat a nonlinear diffusion problem which involves conditions specified on some moving boundary. Since the dynamic behavior and the diffusion process are coupled, there is an even greater degree of complexity in the analysis.

The net inflow of gas which has just been described has been called "rectification of mass". An approximate treatment of the effect has been given by Blake.¹ Blake estimated the effect by a quasi-static solution of the diffusion problem which included only the effect of the area change of the bubble as described in the intuitive argument just given. The present paper gives a complete analysis based on a linearization procedure. The convection effects in the diffusion process are included in the present treatment, and it is found that these convection effects are of the greatest importance in determining the net result.

It will be shown that the radius of a bubble grows slowly due to rectification, and becomes asymptotically proportional to $t^{1/2}$. On the basis of this result, one might expect that in a liquid saturated with dissolved gas a bubble in an oscillating pressure field would grow indefinitely.

1

F.G. Blake, Technical Memorandum No. 12, Acoustics Research Lab., Harvard University (1949).

Such indefinite growth is not observed, and the question therefore is raised regarding the stability of the spherical shape of an oscillating bubble. A brief discussion of this stability question is given in the last section of this paper which shows that there is an upper stability limit for the gas bubble.

Formulation of the Problem

For the case of present interest for which the effect of diffusion is of primary concern, the dynamic problem can be by-passed if one prescribes the oscillating pressure inside the gas bubble rather than the pressure in the liquid at infinity. This procedure is indeed not so arbitrary as might appear for, when the steady state is attained, the pressure inside the bubble behaves in the linearized approximation in essentially the same manner as the external applied pressure except for a phase difference and an unimportant modification of amplitude.*

The pressure within the gas bubble will be taken to be uniform throughout the interior and will be denoted by $P(t)$. $P(t)$ is prescribed in the following manner

$$P(t) = P_0 (1 + \epsilon \sin \omega t), \quad (1)$$

where it is assumed that $\epsilon \ll 1$ so that the linearization procedure may be carried out. It is also assumed that the gas inside the bubble behaves isothermally during expansion and compression. It follows that

$$R(t) = R_0 (1 + \delta \sin \omega t) + O(\delta^2), \quad (2)$$

where

$$-3\delta = \epsilon, \quad ,$$

R is the radius of the bubble at time t , and R_0 is the equilibrium radius corresponding to P_0 .

A more general assumption regarding the thermodynamic behavior of the bubble leads to a phase difference between $(P - P_0)$ and $(R - R_0)$. This possibility is not considered since no essentially new feature is introduced by this complication.

* The dynamic problem is considered in detail by the authors elsewhere.

The amount of gas flowing into the bubble in a time interval Δt is

$$\int_{t_0}^{t_0 + \Delta t} dt \int_S D \nabla C \cdot d\mathbf{S} ,$$

where C is the concentration of gas dissolved in the liquid, D is the coefficient of diffusion, and the integration is over the surface S of the bubble wall. When the problem possesses spherical symmetry, as in the present case, this expression simplifies to

$$\int_{t_0}^{t_0 + \Delta t} dt 4\pi D R^2 \left(\frac{\partial C}{\partial r} \right)_{r=R} .$$

The concentration C is a solution of the diffusion equation

$$\frac{\partial C}{\partial t} + \mathbf{q} \cdot \nabla C = D \nabla^2 C , \quad (3)$$

with appropriate initial and boundary conditions. In Eq. (3) \mathbf{q} is the flow velocity of the liquid. For an irrotational flow field in the liquid it is known that

$$\mathbf{q} = \frac{R^2 \dot{R}}{r^3} \mathbf{r} , \quad (4)$$

with $\dot{R} \equiv dR/dt$.

The boundary conditions are specified in the following way. The amount of gas dissolved in the liquid does not change with time at a large distance from the bubble, that is, $C \rightarrow C_\infty$, a constant, as $r \rightarrow \infty$. The dissolved concentration in the liquid at the bubble wall is determined in accordance with Henry's Law which says that the concentration of dissolved gas at constant temperature is proportional to the pressure; it follows that at $r = R$, $C = aP(R)$ where a is a constant characteristic of the liquid-gas combination. Also, since $C = C_\infty$ everywhere when there is no disturbance in the equilibrium situation, one has $aP_0 = C_\infty$. The formulation of the boundary conditions for the solution of Eq. (3) is, thus,

$$C = C_\infty , \quad \text{as } r \rightarrow \infty ; \quad (5)$$

$$C = C_\infty (1 + \epsilon \sin \omega t), \quad \text{at } r = R . \quad (6)$$

The initial condition is specified as follows:

$$C(r, t) = C_{\infty}, \quad \text{for } t \leq 0, \quad \text{and for all } r. \quad (7)$$

Since one is interested in the steady state solution, only the asymptotic solution for large t need be found.

Solution of the Problem

One may define $\theta(r, t)$ as

$$\theta(r, t) = C(r, t) - C_{\infty}.$$

The problem is then reduced to the solution of the equation

$$L(\theta) \equiv \frac{\partial^2(r\theta)}{\partial r^2} - \frac{1}{D} \frac{\partial(r\theta)}{\partial t} = \frac{R^2 \dot{R}}{Dr} \frac{\partial \theta}{\partial r} \equiv g(\theta), \quad (8)$$

with the conditions

$$\theta(r, 0) = \theta(\infty, t) = 0; \quad (9)$$

and

$$\theta(R(t), t) = \epsilon C_{\infty} \sin \omega t = -3\delta C_{\infty} \sin \omega t. \quad (10)$$

A scheme of successive approximations in powers of the small parameter ϵ can be developed, and the leading term that contributes to the rectification of mass will be evaluated explicitly here. This leading term is of order ϵ^2 . The successive approximations may be carried out in two steps. First, one solves the following problem:

$$L(\theta_1) = 0, \quad (11)$$

with

$$\theta_1(r, 0) = \theta_1(\infty, t) = 0, \quad (12)$$

and

$$\theta_1(R(t), t) = -3\delta C_{\infty} \sin \omega t. \quad (13)$$

With this solution as the first approximation for $g(\theta)$, one then carries out the next step of successive approximations by solving the equation

$$L(\theta_2) = g(\theta_1 + \theta_2), \quad (14)$$

with

$$\theta_2(r, 0) = \theta_2(\infty, t) = 0, \quad (15)$$

and

$$\theta_2(R(t), t) = 0. \quad (16)$$

When θ_1 and θ_2 have been obtained in this way, then $\theta = \theta_1 + \theta_2$ will be the solution to the desired order of accuracy. It may be pointed out that to obtain a solution accurate in second order in ϵ (or δ) the bubble boundary is specified with sufficient accuracy as

$$R(t) = R_0(1 + \delta \sin \omega t).$$

To obtain solutions to orders higher than ϵ^2 requires that the expression for $R(t)$ be corrected to the appropriate degree of accuracy. It is easy to see that this scheme of successive approximation, although workable in principle for higher order solutions, becomes quite complicated.

The asymptotic solution of Eqs. (11), (12), and (13) for large t is to the order of ϵ^2 (cf. Appendix I) :

$$\begin{aligned} \theta_1(r, t) = & - \frac{3R_0 C_\infty \delta}{r} \left\{ e^{-(r-R_0)\sqrt{\omega/2D}} \sin \left[\omega t - (r-R_0)\sqrt{\omega/2D} \right] \right. \\ & + \frac{\delta}{2} \left[(1 + R_0\sqrt{\omega/2D}) \operatorname{Erfc} \left(\frac{r-R_0}{\sqrt{4Dt}} \right) - (1 + R_0\sqrt{\omega/2D}) e^{-(r-R_0)\sqrt{\omega/2D}} \cos \left(2\omega t - (r-R_0)\sqrt{\omega/D} \right) \right. \\ & \left. \left. + R_0\sqrt{\omega/2D} e^{-(r-R_0)\sqrt{\omega/D}} \sin \left(2\omega t - (r-R_0)\sqrt{\omega/D} \right) \right] \right\} + O(t^{-3/2}). \quad (17) \end{aligned}$$

For the calculation of the rectification of mass, it is not necessary to evaluate θ_2 explicitly since the rate of gas flow into the bubble is determined by $(\partial \theta_2 / \partial r)_{r=R}$. This quantity may be found by use of Eq. (17) for large t , up to the order ϵ^2 , to be (cf. Appendix II)

$$\left(\frac{\partial \theta_2}{\partial r} \right)_{r=R} = - \frac{1}{R_0} \int_{R_0}^{\infty} g_1(r) dr + O\left(\frac{1}{t^{3/2}} \right) + S, \quad (18)$$

where S consists of sinusoidal terms which do not contribute to the net flow of gas into the bubble over a complete cycle in the order of ϵ^2 . The $g_1(r)$ is given by

$$g_1(r) = 3R_o^4 C_\infty \left(\frac{\omega}{2D}\right) \delta^2 e^{-(r-R_o)\sqrt{\omega/2D}} \left\{ - \left[\frac{1}{r^3} + \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \right] \sin \left[(r-R_o)\sqrt{\omega/2D} \right] + \frac{1}{r^2} \sqrt{\omega/2D} \cos \left[(r-R_o)\sqrt{\omega/2D} \right] \right\}. \quad (19)$$

The integral which occurs in Eq. (18) is not readily evaluated in the general case. However, for the case that $R_o \sqrt{\omega/2D} \gg 1$, i.e., when the diffusion length $\sqrt{D/\omega}$ is small compared with the bubble radius*, one can obtain an asymptotic expression. In this way, one finds (cf. Appendix III)

$$\left(\frac{\partial \theta_2}{\partial r} \right)_{r=R} = C_\infty \delta^2 \left\{ \left[-\frac{3}{2} \sqrt{\frac{\omega}{2D}} + \frac{9}{2R_o} \right] + 0 \left(\frac{1}{R_o^2 \sqrt{\omega/2D}} \right) + 0 \left(\frac{1}{t^{1/2}} \right) + S \right\} + 0(\delta^3). \quad (20)$$

From Eq. (17) one obtains (cf. Appendix I)

$$\begin{aligned} \left(\frac{\partial \theta_1}{\partial r} \right)_{r=R} = & 3R_o C_\infty \delta \left\{ \left[\frac{1}{R_o^2} + \frac{1}{R_o} \sqrt{\frac{\omega}{2D}} \right] \sin \omega t + \frac{1}{R_o} \sqrt{\frac{\omega}{2D}} \cos \omega t \right\} \\ & + 3R_o C_\infty \delta^2 \left\{ \frac{1}{2R_o^2} \left[1 + R_o \sqrt{\frac{\omega}{2D}} \right] - \left[\frac{2}{R_o^2} + \frac{2}{R_o} \sqrt{\frac{\omega}{2D}} \right] \sin^2 \omega t \right. \\ & \left. + 0 \left(\frac{1}{t^{1/2}} \right) + S' \right\} + 0(\delta^3), \quad (21) \end{aligned}$$

where S' contains only sinusoidal terms which will not contribute to rectification up to order ϵ^2 .

The rate of gas flow into the bubble is

* The value of D for air in water at 20°C is approximately $2 \times 10^{-5} \text{ cm}^2/\text{sec}$.

$$J = 4 \pi D R^2 \left[\frac{\partial \theta}{\partial r} \right]_{r=R} = 4 \pi D R_o^2 \left[1 + 2 \delta \sin \omega t + 0(\delta^2) \right] \left[\frac{\partial}{\partial r} (\theta_1 + \theta_2) \right]_{r=R},$$

which by application of Eqs. (20) and (21) becomes, to the order of ϵ^2 ,

$$J = 24 \pi D C_\infty R_o \delta^2 \left[1 + 0 \left(\frac{1}{R_o \sqrt{\omega/2D}} \right) + 0 \left(\frac{1}{t^{1/2}} \right) \right] + S + 0(\delta^2). \quad (22)$$

Thus, the leading term for the average rate of flow of gas into the bubble is

$$\overline{J} = 24 \pi D C_\infty R_o \delta^2. \quad (23)$$

Since

$$\delta = -\frac{\epsilon}{3} = -\frac{1}{3} \frac{P_{\max} - P_o}{P_o} = -\frac{1}{3} \frac{\Delta P}{P_o}$$

one may also write

$$\overline{J} = \frac{8}{3} \pi D C_\infty R_o \left(\frac{\Delta P}{P_o} \right)^2. \quad (24)$$

Thus, when the ratio of the pressure amplitude ΔP to the mean pressure P_o is sufficiently small, the bubble growth by rectification is determined by this leading term.

The mass of gas inside the bubble is

$$m = \frac{4}{3} \pi \rho_g R_o^3. \quad (25)$$

The mean density of the gas, ρ_g , remains essentially unchanged during the slow growth so that

$$\frac{dm}{dt} = 4 \pi \rho_g R_o^2 \frac{dR_o}{dt}. \quad (26)$$

On the other hand, the rate of increase of mass in the bubble by rectification is

$$\frac{dm}{dt} = \overline{J} = \frac{8 \pi}{3} D C_\infty \epsilon^2 R_o \quad (27)$$

so that Eqs. (26) and (27) give

$$\frac{dR_o}{dt} = \frac{2}{3} \frac{D C_\infty \epsilon^2}{\rho_g R_o} \quad (28)$$

It follows that

$$R_o^2 = R_i^2 + \frac{4}{3} \frac{D C_\infty \epsilon^2}{\rho_g} t \quad (29)$$

if one sets $R_o = R_i$ at $t = 0$. Equation (29) may be written alternatively in the form

$$R_o = 2 \left(\frac{D C_\infty}{3 \rho_g} \right)^{1/2} \epsilon (t + t_o)^{1/2} \quad (30)$$

where

$$t_o = \frac{3 \rho_g}{D C_\infty} \left(\frac{R_i}{\epsilon} \right)^2.$$

A measure of the rectification growth rate is the time τ required for a bubble to double its size. From Eq. (29), one obtains for this time

$$\tau = \frac{9 R_o^2 \rho_g}{4 C_\infty D \epsilon^2}.$$

Some numerical values for the case of air in water at 20°C and 1 atm pressure are given in Table I.

Stability of a Spherical Gas Bubble in an Oscillating Pressure Field

The result of the calculation of mass rectification indicates that a spherical gas bubble in a liquid with an oscillating pressure field grows indefinitely. Such a behavior is not observed experimentally so that the question naturally arises concerning the stability of the spherical bubble in an oscillating pressure field. The general relations which determine the stability of this spherical flow have been discussed elsewhere² and have

² M.S. Plesset, J. Appl. Phys., 25, 96 (1954).

TABLE I

Time Required for Air Bubbles in Water at 20°C, 1 atm,
to Double in Size by Mass Rectification

Initial Radius $R_i(\text{cm})$	Relative Pressure Amplitude $\epsilon = \frac{P_{\text{max}} - P_o}{P_o}$	Doubling Time $\tau(\text{sec})$
10^{-1}	0.25	1.1×10^6
10^{-1}	0.10	6.7×10^6
10^{-1}	0.01	6.7×10^8
10^{-2}	0.25	1.1×10^4
10^{-2}	0.10	6.7×10^4
10^{-2}	0.01	6.7×10^6
10^{-3}	0.25	1.1×10^2
10^{-3}	0.10	6.7×10^2
10^{-3}	0.01	6.7×10^4

been applied to the specific case of the growth and collapse of vapor bubbles.³ The treatment given here will apply the basic relations derived in Refs. 2 and 3 to the problem of the oscillating gas bubble.

It is apparent that so far as the effect of the rectification of mass is concerned, the growth of a gas bubble is very slow. Therefore, the stability considerations may be applied to the case in which the mean radius of the bubble remains essentially constant in time.

Let the bubble boundary be distorted from a spherical surface of radius R to a surface with radius vector of magnitude r_S . Then one may write

$$r_S = R + \sum_n a_n Y_n, \quad (31)$$

where Y_n is a spherical harmonic of degree n and the a_n 's are functions of the time to be determined. The growth or decay of $a_n(t)$ from a small initial value determines whether the spherical shape is unstable or stable. When a linearized perturbation procedure under the assumption that

$$|a_n(t)| \ll R(t)$$

is applied to the case of two immiscible, incompressible, inviscid fluids separated by a spherical interface, one finds² that the a_n 's are independent of each other and that they satisfy the following differential equation:

$$\frac{d^2 a_n}{dt^2} + \frac{3}{R} \frac{dR}{dt} \frac{da_n}{dt} - A a_n = 0. \quad (32)$$

The function A in Eq. (32) is given by

$$A = \frac{[n(n-1)\rho_2 - (n+1)(n+2)\rho_1] \frac{d^2 R}{dt^2} - (n-1)n(n+1)(n+2)\sigma/R^2}{[n\rho_2 + (n+1)\rho_1] R}, \quad (33)$$

where ρ_1 is the density of the fluid inside the sphere, ρ_2 is the density of the fluid outside the sphere, and σ is the surface tension constant.

³ M.S. Plesset and T.P. Mitchell, *Quart. Appl. Math.*, 8, 419 (1956)

Although the stability of the spherical shape for a small distortion may be inferred from the decay of $a_n(t)$ with time, strictly speaking the instability inferred from a growth of $a_n(t)$ with time is a reasonable conjecture rather than a necessary consequence because of the linearization process.

For the case of a gas bubble, the gas density ρ_1 may be neglected in comparison with the liquid density ρ_2 . Then A becomes

$$A = \frac{(n-1)}{R} \frac{d^2 R}{dt^2} - (n-1)(n+1)(n+2) \frac{\sigma}{\rho R^3}, \quad (34)$$

where $\rho = \rho_2$ is the liquid density. If one writes

$$b_n = R^{2/3} a_n, \quad (35)$$

then Eq. (32) is transformed into

$$\frac{d^2 b_n}{dt^2} + G(t) b_n = 0, \quad (36)$$

where

$$G_n = (n-1)(n+1)(n+2) \frac{\sigma}{\rho R^3} - \frac{3}{4R^2} \left(\frac{dR}{dt} \right)^2 - \frac{(n+1/2)}{R} \frac{d^2 R}{dt^2}. \quad (37)$$

The radius of the undisturbed bubble is determined as a function of time by the familiar equation

$$R \frac{d^2 R}{dt^2} + \frac{3}{2} \left(\frac{dR}{dt} \right)^2 = \frac{1}{\rho} (P_i - P_\infty - \frac{2\sigma}{R}), \quad (38)$$

where P_i is the pressure inside the bubble, and P_∞ is the pressure at a distance from the bubble which in the present case may be expressed as

$$P_\infty = P_0 (1 + \epsilon \sin \omega t). \quad (39)$$

When ϵ is small compared with unity, a linearized calculation gives the following solution for R :

$$R = R_0 \left[1 + \delta \sin(\omega t + \phi) \right], \quad (40)$$

where δ is of the same order of magnitude as ϵ and \emptyset is a constant phase shift, which for convenience may be put equal to zero. Then $G(t)$ may be expressed as

$$\begin{aligned} G(t) = & (n-1)(n+1)(n+2) \frac{\sigma}{\rho R_o^3} \left[1 - 3\delta \sin \omega t + O(\delta^2) \right] \\ & - \frac{3}{4} \delta^2 \omega^2 \left[1 - 2\delta \sin \omega t + O(\delta^2) \right] \cos^2 \omega t \\ & + (n + \frac{1}{2}) \delta \omega^2 \sin \omega t \left[1 - \delta \sin \omega t + O(\delta^2) \right]. \end{aligned} \quad (41)$$

With this expression for $G(t)$, the differential equation (36) can be recognized as belonging to the kind of equations known as Hill's equation. With $\delta \ll 1$ one may retain the leading terms in the expression for $G(t)$ which then takes the form

$$G(t) = \alpha + \beta \sin \omega t, \quad (42)$$

where

$$\alpha = (n-1)(n+1)(n+2) \frac{\sigma}{\rho R_o^3} + O(\delta^2), \quad (43)$$

and

$$\beta = \delta \left[(n + \frac{1}{2}) \omega^2 - (n-1)(n+1)(n+2) \frac{3\sigma}{\rho R_o^3} \right] + O(\delta^2) \quad (44)$$

Equation (36) is then just the Mathieu equation.

The stability theory of solutions of the Mathieu equation is well known.⁴ Relations between the parameters $n, \sigma, \rho, \delta, \omega$, and R_o may be obtained to determine the region of stability or instability of the solutions. More specifically, one may determine the critical value of R_o which is the transition value between stability and instability for given values of σ, ρ, δ and ω . Without going into the details of determining the exact stability conditions, one may indicate how the critical radius is determined with the aid of the stability chart for the Mathieu equation.⁴ The solution is essentially unstable if $G < 0$. In applying this criterion to Eq. (43) one

⁴ See, for example, N.W. McLachlan, "Theory and Applications of Mathieu Functions", Clarendon Press, Oxford (1947).

must keep in mind that the values of n of interest do not include $n=1$ since this value of n corresponds not to a distortion of the spherical shape but to a translation of the entire bubble.

One may see from the behavior of α and β , or from examination of the stability chart, that the greater n the greater is the limit of stability. Therefore, for the determination of the critical radius, it is sufficient to consider the case $n=2$ only. In this case Eq. (42) becomes

$$G(t) = \frac{12\sigma}{\rho R_o^3} + \delta \left(\frac{5}{2} \omega^2 - \frac{36\sigma}{\rho R_o^3} \right) \sin \omega t . \quad (45)$$

An order of magnitude criterion of stability is thus

$$\frac{12\sigma}{\rho R_o^3} \gtrsim \frac{5}{2} \delta \omega^2 . \quad (46)$$

From Eq. (46), one gets

$$(R_o)_{cr} \sim \left(\frac{24\sigma}{5\rho\delta\omega^2} \right)^{1/3} \quad (47)$$

The solution is stable only if the mean radius R_o is less than $(R_o)_{cr}$.

This general result may be illustrated by considering the particular case of an air bubble in water. One then has $\sigma = 73.5$ dyne/cm and $\rho = 1$ gm/cm³. If one now takes the example of $\delta = 10^{-2}$ and $\omega = 10^4$ /sec, then

$$(R_o)_{cr} \sim 10^{-1} \text{ cm} .$$

This value is very reasonable in view of the experimental observations with sonic and ultrasonic pressure oscillations in water. If the critical radius were found to be appreciably larger than this value, the process of rectification would lead to the eventual formation of large air bubbles in water subject to pressure oscillations. Experimental observations do not show the appearance of such large bubbles.

Appendix I

To solve

$$\frac{\partial}{\partial t} (r \theta_1) = D \frac{\partial^2}{\partial r^2} (r \theta_1) , \quad (\text{I } 1)$$

with

$$\theta_1(r, 0) = \theta_1(\infty, t) = 0 , \quad (\text{I } 2)$$

and

$$\theta_1(R(t), t) = -3 C_{\infty} \delta \sin \omega t , \quad (\text{I } 3)$$

up to $O(\delta^2)$, we note that

$$\theta_1(R(t), t) = \theta_1(R_o, t) + (R - R_o) \left(\frac{\partial \theta_1}{\partial r} \right)_{r=R_o} + \dots ,$$

or

$$\theta_1(R_o, t) = -3 C_{\infty} \delta \sin \omega t - R_o \delta \sin \omega t \left(\frac{\partial \theta_1}{\partial r} \right)_{r=R_o} - \dots , \quad (\text{I } 4)$$

We may ignore the remaining terms, since they are of the order of δ^3 .

Now let us solve first the equation

$$\frac{\partial}{\partial t} (r \theta_o) = D \frac{\partial^2}{\partial r^2} (r \theta_o) , \quad (\text{I } 5)$$

with

$$\theta_o(r, 0) = \theta_o(\infty, t) = 0 , \quad (\text{I } 6)$$

and

$$\theta_o(R_o, t) = -3 C_{\infty} \delta \sin \omega t . \quad (\text{I } 7)$$

Denote

$$v_o(r; s) = \mathcal{L}\{r \theta_o\} = \int_0^{\infty} r \theta_o(r, t) e^{-st} dt . \quad (\text{I } 8)$$

Then the transformed equation and conditions become

$$\frac{d^2 v_o}{dr^2} = \frac{s}{D} v_o , \quad (I\ 9)$$

with

$$\lim_{r \rightarrow \infty} \frac{v_o}{r} = 0 ,$$

and

$$v_o(R_o; s) = -3 R_o C_\infty \delta \frac{\omega}{s^2 + \omega^2} . \quad (I\ 10)$$

Thus

$$v(r; s) = - \frac{3 R_o C_\infty \omega \delta}{s^2 + \omega^2} e^{-(r-R_o)\sqrt{s/D}} . \quad (I\ 11)$$

Using the inversion formula, we obtain

$$r \theta_o(r, t) = - \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \frac{3 R_o C_\infty \omega \delta e^{-(r-R_o)\sqrt{s/D}}}{s^2 + \omega^2} e^{st} ds . \quad (I\ 12)$$

Thus for large t , we have

$$\theta_o(r, t) = - \frac{3 R_o C_\infty \delta}{r} e^{-(r-R_o)\sqrt{\omega/2D}} \sin \left[\omega t - (r-R_o)\sqrt{\frac{\omega}{2D}} \right] + O(t^{-3/2}) ; \quad (I\ 13)$$

and

$$\begin{aligned} \frac{\partial \theta_o}{\partial r} = 3 R_o C_\infty \delta e^{-(r-R_o)\sqrt{\omega/2D}} & \left\{ \left[\frac{1}{r^2} + \frac{1}{r} \sqrt{\frac{\omega}{2D}} \right] \sin \left[\omega t - (r-R_o)\sqrt{\frac{\omega}{2D}} \right] \right. \\ & \left. + \frac{1}{r} \sqrt{\frac{\omega}{2D}} \cos \left[\omega t - (r-R_o)\sqrt{\frac{\omega}{2D}} \right] \right\} + O(t^{-3/2}) . \end{aligned} \quad (I\ 14)$$

Neglecting terms of the order of $t^{-3/2}$, we thus have

$$\left(\frac{\partial \theta_o}{\partial r} \right)_{r=R_o} = \frac{3 C_\infty \delta}{R_o} \left[\left(1 + R_o \sqrt{\frac{\omega}{2D}} \right) \sin \omega t + R_o \sqrt{\frac{\omega}{2D}} \cos \omega t \right] . \quad (I\ 15)$$

Now θ_1 will be solved by putting in (4) $\left(\frac{\partial \theta_o}{\partial r} \right)_{r=R_o}$ in place of $\left(\frac{\partial \theta_1}{\partial r} \right)_{r=R_o}$.

Then

$$R_o \theta_1(R_o, t) = -3 R_o C_\infty \delta \left\{ \sin \omega t + \delta \left[\left(1 + R_o \sqrt{\frac{\omega}{2D}} \right) \sin^2 \omega t + \frac{R_o}{2} \sqrt{\frac{\omega}{2D}} \sin 2\omega t \right] \right\} . \quad (I 16)$$

Now let $v_1(r; s) = \mathcal{L}\{r \theta_1\}$; it is easy to see that

$$v_1(r; s) = v_1(R_o; s) e^{-(r-R_o)\sqrt{s/D}} , \quad (I 17)$$

where

$$v_1(R_o; s) = -3 R_o C_\infty \delta \left\{ \frac{\omega}{s^2 + \omega^2} + \frac{\delta}{2} \left[\left(1 + R_o \sqrt{\frac{\omega}{2D}} \right) \left(\frac{1}{s} - \frac{s}{s^2 + 4\omega^2} \right) + R_o \sqrt{\frac{\omega}{2D}} \frac{2\omega}{s^2 + 4\omega^2} \right] \right\} . \quad (I 18)$$

Thus we obtain from the inversion formula asymptotically for large t :

$$\begin{aligned} \theta_1(r, t) = & - \frac{3 R_o C_\infty \delta}{r} \left\{ e^{-(r-R_o)\sqrt{\omega/2D}} \sin \left[\omega t - (r-R_o) \sqrt{\frac{\omega}{2D}} \right] \right. \\ & + \frac{\delta}{2} \left[\left(1 + R_o \sqrt{\frac{\omega}{2D}} \right) \operatorname{Erfc} \left(\frac{r-R_o}{\sqrt{4Dt}} \right) - \left(1 + R_o \sqrt{\frac{\omega}{2D}} \right) e^{-(r-R_o)\sqrt{\frac{\omega}{2D}}} \cos \left[2\omega t - (r-R_o) \sqrt{\frac{\omega}{2D}} \right] \right. \\ & \left. \left. + R_o \sqrt{\frac{\omega}{2D}} e^{-(r-R_o)\sqrt{\frac{\omega}{2D}}} \sin \left[2\omega t - (r-R_o) \sqrt{\frac{\omega}{2D}} \right] \right] \right\} + O(t^{-3/2}) . \quad (I 19) \end{aligned}$$

Now

$$\left(\frac{\partial \theta_1}{\partial r} \right)_{r=R} = \left(\frac{\partial \theta_1}{\partial r} \right)_{r=R_o} + (R-R_o) \left(\frac{\partial^2 \theta_1}{\partial r^2} \right)_{r=R_o} + O(\delta^3) . \quad (I 20)$$

From (19), we have

$$\begin{aligned}
\frac{\partial \theta_1}{\partial r} = & 3 R_o C_\infty \delta \left\{ \left[\frac{1}{r^2} + \frac{1}{r} \sqrt{\frac{\omega}{2D}} \right] \sin \left[\omega t - (r - R_o) \sqrt{\frac{\omega}{2D}} \right] \right. \\
& + \left. \frac{1}{r} \sqrt{\frac{\omega}{2D}} \cos \left[\omega t - (r - R_o) \sqrt{\frac{\omega}{2D}} \right] \right\} e^{-(r - R_o) \sqrt{\omega/2D}} \\
& + \frac{3}{2} R_o C_\infty \delta^2 \left\{ \left[1 + R_o \sqrt{\frac{\omega}{2D}} \right] \left[\frac{1}{r^2} \operatorname{Erfc} \left(\frac{r - R_o}{\sqrt{4Dt}} \right) \right. \right. \\
& + \left. \left. \frac{1}{r \sqrt{\pi Dt}} e^{-(r - R_o)^2/4Dt} \right] + S \right\} , \tag{I 21}
\end{aligned}$$

where S denotes those sinusoidal terms which will not contribute to the rectification up to the second order.

Also

$$\begin{aligned}
\frac{\partial^2 \theta_1}{\partial r^2} = & -3 R_o C_\infty \delta \left\{ \left[\frac{2}{r^3} + \frac{2}{r^2} \sqrt{\frac{\omega}{2D}} \right] \sin \left[\omega t - (r - R_o) \sqrt{\frac{\omega}{2D}} \right] \right. \\
& + \left. \left[\frac{2}{r^2} \sqrt{\frac{\omega}{2D}} + \frac{2}{r} \frac{\omega}{2D} \right] \cos \left[\omega t - (r - R_o) \sqrt{\frac{\omega}{2D}} \right] \right\} e^{-(r - R_o) \sqrt{\omega/2D}} \\
& + O(\delta^2) . \tag{I 22}
\end{aligned}$$

Thus from (20), since $R - R_o = R_o \delta \sin \omega t$, we have

$$\begin{aligned}
\left(\frac{\partial \theta_1}{\partial r} \right)_{r=R} = & 3 R_o C_\infty \delta \left[\left(\frac{1}{R_o^2} + \frac{1}{R_o} \sqrt{\frac{\omega}{2D}} \right) \sin \omega t + \frac{1}{R_o} \sqrt{\frac{\omega}{2D}} \cos \omega t \right] \\
& + 3 R_o C_\infty \delta^2 \left[\left(1 + R_o \sqrt{\frac{\omega}{2D}} \right) \left(\frac{1}{2 R_o^2} + \frac{1}{2 R_o \sqrt{\pi Dt}} + \frac{2}{R_o^2} \sin^2 \omega t \right) + S \right] \\
& + O(t^{-3/2}) + O(\delta^3) . \tag{I 23}
\end{aligned}$$

Appendix II

We want to solve the following equation :

$$\frac{\partial^2}{\partial r^2} (r \theta_2) - \frac{1}{D} \frac{\partial}{\partial t} (r \theta_2) = g(r, t) , \quad (\text{II } 1)$$

where $g(r, t) = \frac{R^2 \dot{R}}{D r} \frac{\partial \theta_1}{\partial r}$ to the order of δ^2 . From the result in Appendix I, since $\dot{R} = \delta R_0 \omega \cos \omega t$ we have

$$g(r, t) = \frac{3 R_0^4 C_\infty \omega \delta^2}{D r} e^{-(r-R_0) \sqrt{\frac{\omega}{2D}}} \left\{ \left[\frac{1}{r^2} + \frac{1}{r} \sqrt{\frac{\omega}{2D}} \right] \sin \left[\omega t - (r-R_0) \sqrt{\frac{\omega}{2D}} \right] + \frac{1}{r} \sqrt{\frac{\omega}{2D}} \cos \left[\omega t - (r-R_0) \sqrt{\frac{\omega}{2D}} \right] \right\} \cos \omega t + O(\delta^3) . \quad (\text{II } 2)$$

The initial and boundary conditions, up to the same order, are

$$\theta_2(r, 0) = \theta_2(\infty, t) = 0 , \quad (\text{II } 3)$$

and

$$\theta_2(R_0, t) = 0 . \quad (\text{II } 4)$$

Apply Laplace Transformation, and let $\phi = \mathcal{L}\{\theta_2\}$, also put

$$h(r; s) = \mathcal{L}\{g(r, t)\} ; \quad (\text{II } 5)$$

then it may be verified that :

$$\begin{aligned} \phi(r; s) = & - \frac{1}{2r} \sqrt{\frac{D}{s}} \left[e^{-(r-R_0) \sqrt{\frac{s}{D}}} \int_{R_0}^r h(x; s) e^{(x-R_0) \sqrt{\frac{s}{D}}} dx \right. \\ & + e^{(r-R_0) \sqrt{\frac{s}{D}}} \int_r^\infty h(x; s) e^{-(x-R_0) \sqrt{\frac{s}{D}}} dx \\ & \left. - e^{-(r-R_0) \sqrt{\frac{s}{D}}} \int_{R_0}^\infty h(x; s) e^{-(x-R_0) \sqrt{\frac{s}{D}}} dx \right] . \end{aligned} \quad (\text{II } 6)$$

Hence

$$\begin{aligned}
 \frac{d\phi}{dr} = & -\frac{1}{2} \sqrt{\frac{D}{s}} \left[\left(-\frac{1}{r^2} - \frac{1}{r} \sqrt{\frac{s}{D}} \right) e^{-(r-R_0)} \sqrt{\frac{s}{D}} \int_{R_0}^r h(x; s) e^{(x-R_0)} \sqrt{\frac{s}{D}} dx \right. \\
 & + \left(-\frac{1}{r^2} + \frac{1}{r} \sqrt{\frac{s}{D}} \right) e^{(r-R_0)} \sqrt{\frac{s}{D}} \int_r^\infty h(x; s) e^{-(x-R_0)} \sqrt{\frac{s}{D}} dx \\
 & \left. + \left(\frac{1}{r^2} + \frac{1}{r} \sqrt{\frac{s}{D}} \right) e^{-(r-R_0)} \sqrt{\frac{s}{D}} \int_{R_0}^\infty h(x; s) e^{-(x-R_0)} \sqrt{\frac{s}{D}} dx \right] . \quad (\text{II } 7)
 \end{aligned}$$

Thus

$$\left(\frac{d\phi}{dr} \right)_{r=R_0} = -\frac{1}{R_0} \int_{R_0}^\infty h(x; s) e^{-(x-R_0)} \sqrt{\frac{s}{D}} dx . \quad (\text{II } 8)$$

Now let us rewrite the expression of $g(r, t)$ in (2). Then we have

$$\begin{aligned}
 g(r, t) = & 3R_0^4 C_\infty \left(\frac{\omega}{2D} \right) \delta^2 \left\{ \left[\left(\frac{1}{r^3} + \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \right) \cos(r-R_0) \sqrt{\frac{\omega}{2D}} \right. \right. \\
 & + \left. \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \sin(r-R_0) \sqrt{\frac{\omega}{2D}} \right] \sin 2\omega t + \left[\left(-\frac{1}{r^3} - \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \right) \sin(r-R_0) \sqrt{\frac{\omega}{2D}} \right. \\
 & + \left. \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \cos(r-R_0) \sqrt{\frac{\omega}{2D}} \right] \left[1 + \cos 2\omega t \right] \left. \right\} e^{-(r-R_0)} \sqrt{\frac{\omega}{2D}} \\
 & + 0(\delta^3) . \quad (\text{II } 9)
 \end{aligned}$$

From (8) it is fairly obvious that excluding those terms which at most contribute to the rectification of the order of $0(\delta^3)$ and $0(t^{-3/2})$ the relevant term in $g(r, t)$ is just

$$\begin{aligned}
 g_1(r) = & 3R_0^4 C_\infty \left(\frac{\omega}{2D} \right) \delta^2 e^{-(r-R_0)} \sqrt{\frac{\omega}{2D}} \left[\left(-\frac{1}{r^3} - \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \right) \sin(r-R_0) \sqrt{\frac{\omega}{2D}} \right. \\
 & \left. + \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \cos(r-R_0) \sqrt{\frac{\omega}{2D}} \right] . \quad (\text{II } 10)
 \end{aligned}$$

As $h_1(r; s) = \mathcal{L} \{ g_1(r) \} = \frac{1}{s} g_1(r)$, it follows that

$$\left(\frac{\partial \theta_2}{\partial r} \right)_{r=R_0} = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} ds \left[-\frac{1}{R_0 s} e^{st} \int_{R_0}^{\infty} e^{-(x-R_0)\sqrt{s/D}} g_1(x) dx \right] + T \quad (\text{II } 11)$$

where T denotes irrelevant terms. As we are only interested in the behavior of the solution for large t , we thus expand $e^{-(x-R_0)\sqrt{s/D}}$ in ascending powers of $s^{1/2}$, and obtain:

$$\left(\frac{\partial \theta_2}{\partial r} \right)_{r=R_0} = -\frac{1}{R_0} \left[\int_{R_0}^{\infty} g_1(x) dx - \frac{1}{\sqrt{\pi D t}} \int_{R_0}^{\infty} (x-R_0) g_1(x) dx \right] + O(t^{-3/2}). \quad (\text{II } 12)$$

Since

$$r g_1(r) = 3 R_0^4 C_{\infty} \left(\frac{\omega}{2D} \right) \delta^2 \frac{d}{dr} \left[\frac{1}{r} e^{-(r-R_0)\sqrt{\frac{\omega}{2D}}} \sin(r-R_0) \sqrt{\frac{\omega}{2D}} \right],$$

therefore

$$\int_{R_0}^{\infty} x g_1(x) dx = 0;$$

and this leads to the result that

$$\left(\frac{\partial \theta_2}{\partial r} \right)_{r=R_0} = -\frac{1}{R_0} \left(1 + \frac{R_0}{\sqrt{\pi D t}} \right) \int_{R_0}^{\infty} g_1(x) dx + O(t^{-3/2}). \quad (\text{II } 13)$$

We may observe that $\left(\frac{\partial \theta_2}{\partial r} \right)_{r=R_0} = O(\delta^2)$. Hence, up to this order we have

$$\begin{aligned} \left(\frac{\partial \theta_2}{\partial r} \right)_{r=R} &\approx \left(\frac{\partial \theta_2}{\partial r} \right)_{r=R_0} = -\frac{1}{R_0} \left(1 + \frac{R_0}{\sqrt{\pi D t}} \right) \int_{R_0}^{\infty} g_1(x) dx + O(t^{-3/2}), \\ &= -3 R_0^3 C_{\infty} \left(\frac{\omega}{2D} \right) \delta^2 \left(1 + \frac{R_0}{\sqrt{\pi D t}} \right) I_1 + O(t^{-3/2}), \end{aligned} \quad (\text{II } 14)$$

where

$$\begin{aligned}
 I_1 = \int_{R_o}^{\infty} e^{-(r-R_o)\sqrt{\frac{\omega}{2D}}} & \left[\left(-\frac{1}{r^3} - \frac{1}{r^2}\sqrt{\frac{\omega}{2D}} \right) \sin(r-R_o)\sqrt{\frac{\omega}{2D}} \right. \\
 & \left. + \frac{1}{r^2}\sqrt{\frac{\omega}{2D}} \cos(r-R_o)\sqrt{\frac{\omega}{2D}} \right] dr .
 \end{aligned}
 \tag{II 15}$$

Appendix III

To evaluate the integral

$$I_1 = \int_{R_0}^{\infty} \left[\left(-\frac{1}{r^3} - \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \right) \sin(r-R_0) \sqrt{\frac{\omega}{2D}} + \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \cos(r-R_0) \sqrt{\frac{\omega}{2D}} \right] e^{-(r-R_0) \sqrt{\frac{\omega}{2D}}} dr, \quad (\text{III } 1)$$

let us note that

$$\begin{aligned} \frac{d}{dr} \left[\frac{1}{r^2} e^{-(r-R_0) \sqrt{\frac{\omega}{2D}}} \sin(r-R_0) \sqrt{\frac{\omega}{2D}} \right] \\ = \left[\left(-\frac{2}{r^3} - \frac{1}{r^2} \right) \sin(r-R_0) \sqrt{\frac{\omega}{2D}} + \frac{1}{r^2} \sqrt{\frac{\omega}{2D}} \cos(r-R_0) \sqrt{\frac{\omega}{2D}} \right] e^{-(r-R_0) \sqrt{\frac{\omega}{2D}}} , \\ = -\frac{1}{r^3} e^{-(r-R_0) \sqrt{\frac{\omega}{2D}}} \sin(r-R_0) \sqrt{\frac{\omega}{2D}} + i_1, \quad (\text{III } 2) \end{aligned}$$

where i_1 is the integrand in I_1 . From (2), we may thus write

$$I_1 = \int_{R_0}^{\infty} \frac{1}{r^3} e^{-(r-R_0) \sqrt{\frac{\omega}{2D}}} \sin(r-R_0) \sqrt{\frac{\omega}{2D}} dr. \quad (\text{III } 3)$$

After changing variables, we have

$$\begin{aligned} I_1 &= \int_0^{\infty} \frac{1}{(x+R_0)^3} e^{-x \sqrt{\omega/2D}} \sin \sqrt{\frac{\omega}{2D}} x dx, \\ &= \text{Im} \left[\int_0^{\infty} \frac{1}{(x+R_0)^3} e^{-\sqrt{\omega/2D} (x-ix)} dx \right]. \quad (\text{III } 4) \end{aligned}$$

Now let $y = \sqrt{2} e^{-i\pi/4} x = (1-i)x$, then apply Cauchy's Theorem to transform the integral along the real axis of the new coordinate system, and we then obtain

$$I_1 = \text{Im} \left[\frac{e^{i\pi/4}}{\sqrt{2}} \int_0^\infty \frac{e^{-\sqrt{\frac{\omega}{2D}} y}}{\left(R_0 + \frac{e^{i\pi/4}}{\sqrt{2}} y \right)^3} dy \right]. \quad (\text{III } 5)$$

For the case that $R_0 \sqrt{\omega/2D} \gg 1$, we may apply Watson's Lemma, and get

$$\begin{aligned} I_1 &= \text{Im} \left[\frac{e^{i\pi/4}}{\sqrt{2} R_0^3} \left(\frac{1}{\sqrt{\frac{\omega}{2D}}} - \frac{3}{\sqrt{2}} e^{i\pi/4} \frac{1}{R_0 \frac{\omega}{2D}} \right) \right] + 0 \left(\frac{1}{R_0^5 \left(\frac{\omega}{2D} \right)^{3/2}} \right), \\ &= \frac{1}{2 R_0^2} \left[\left(\frac{1}{R_0 \sqrt{\frac{\omega}{2D}}} - \frac{3}{R_0^2 \frac{\omega}{2D}} \right) + 0 \left(\frac{1}{\left(R_0 \sqrt{\frac{\omega}{2D}} \right)^3} \right) \right]. \end{aligned} \quad (\text{III } 6)$$

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